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(54) Abstract Title
Reactor with a heat exchanger structure

(57) A reactor for catalytically converting a flow of operating substance has a heat exchanger structure in the form of a bank of tubes 4 with at least a first and, in heat contact therewith, a second heat exchanger chamber, one of which contains reaction chamber with a conversion catalyst for converting the flow of operating substance delivered thereto, heat conductive, corrugated ribs 3 coated with the conversion catalyst being mounted in the interior 8 of reaction chamber. The flat tube block may be inserted in a housing and the heat exchanger chamber outside the flat tubes contain flow passages running in the longitudinal direction of its associated corrugated ribs 9, the housing having respective connecting structures for delivering and discharging heat exchange medium passing through the heat exchange chamber outside the flat tubes and the flow of operating substance directed through the reaction chamber. The flat tubes may be made up of two tube half-shells, which are prefabricated to have a widening cross section towards the exterior and which are joined together in abutment.

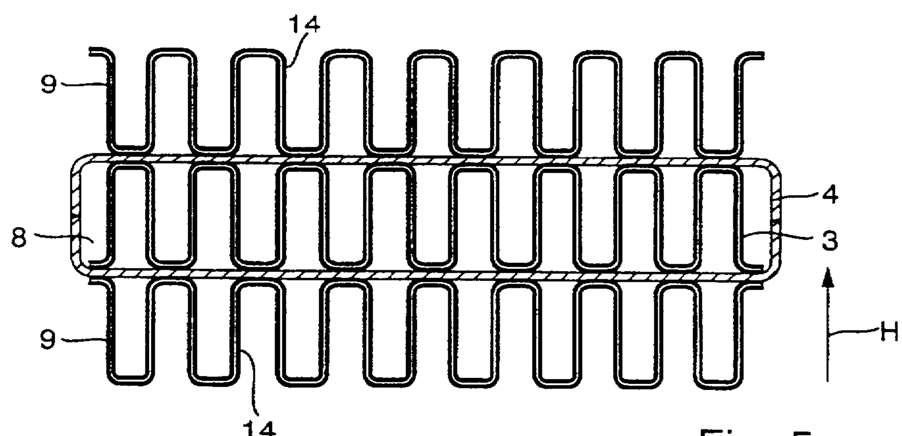

Fig. 5

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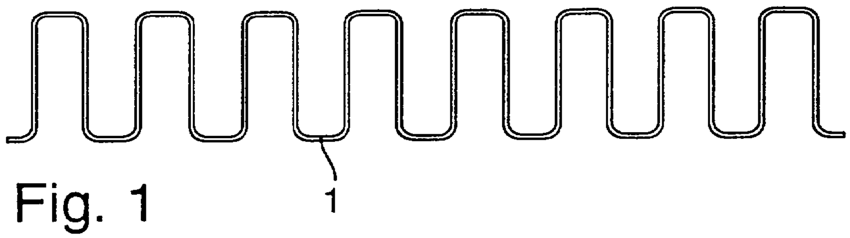


Fig. 1

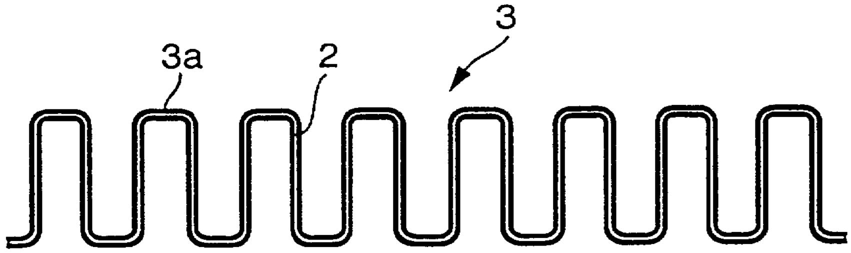


Fig. 2

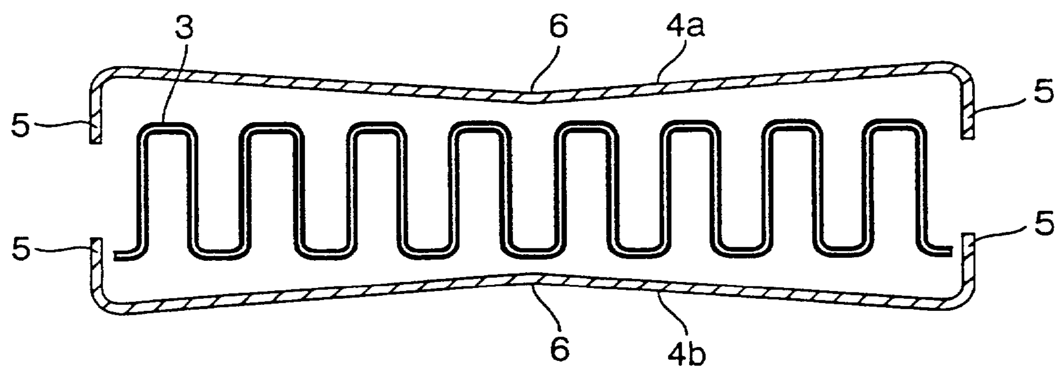


Fig. 3

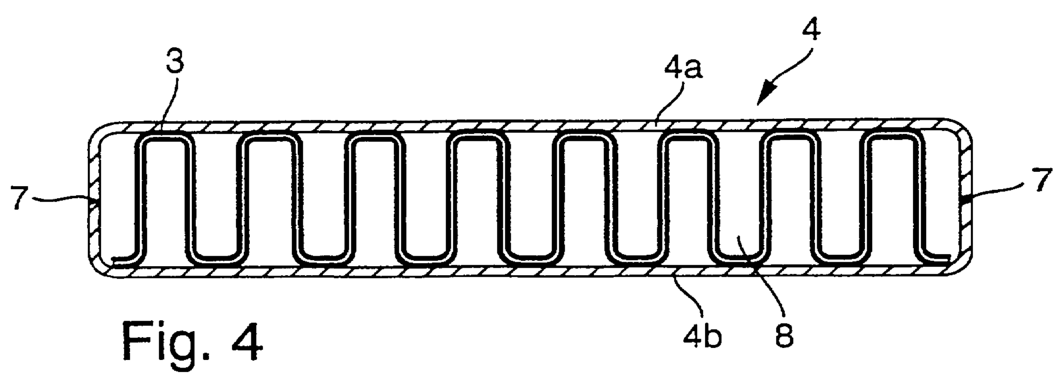


Fig. 4

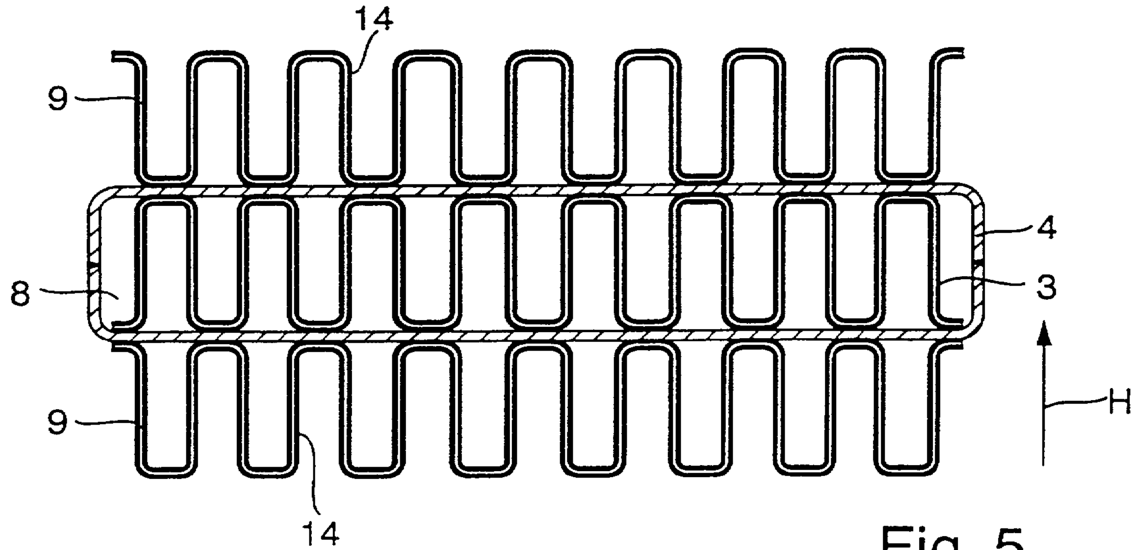


Fig. 5

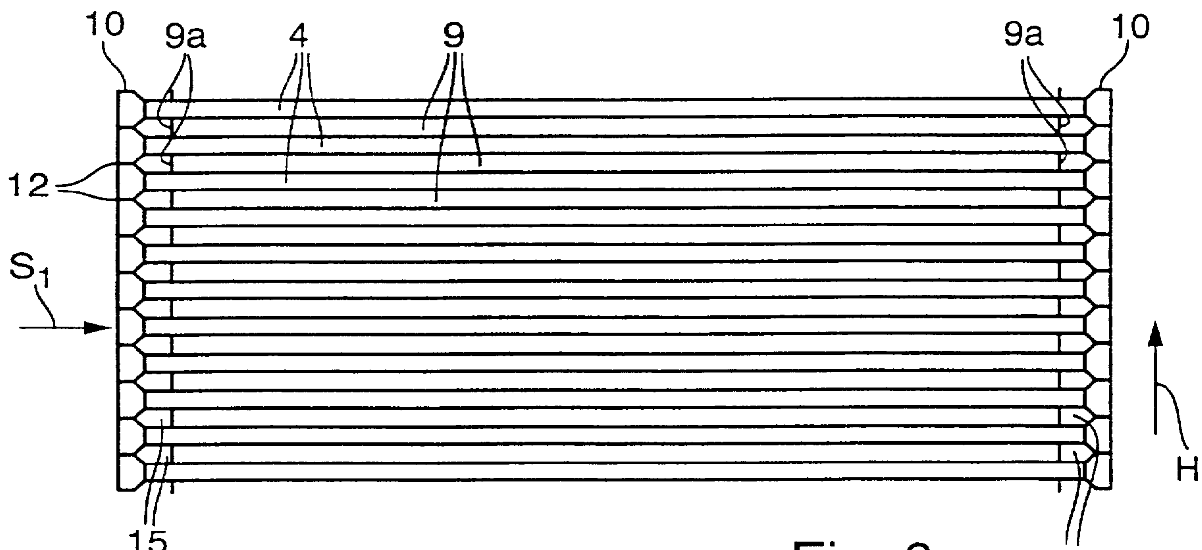


Fig. 6

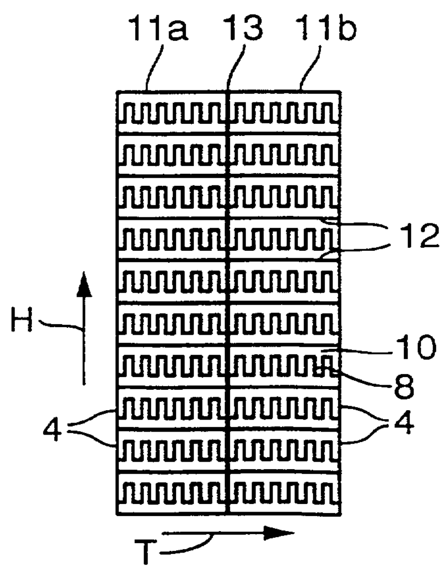
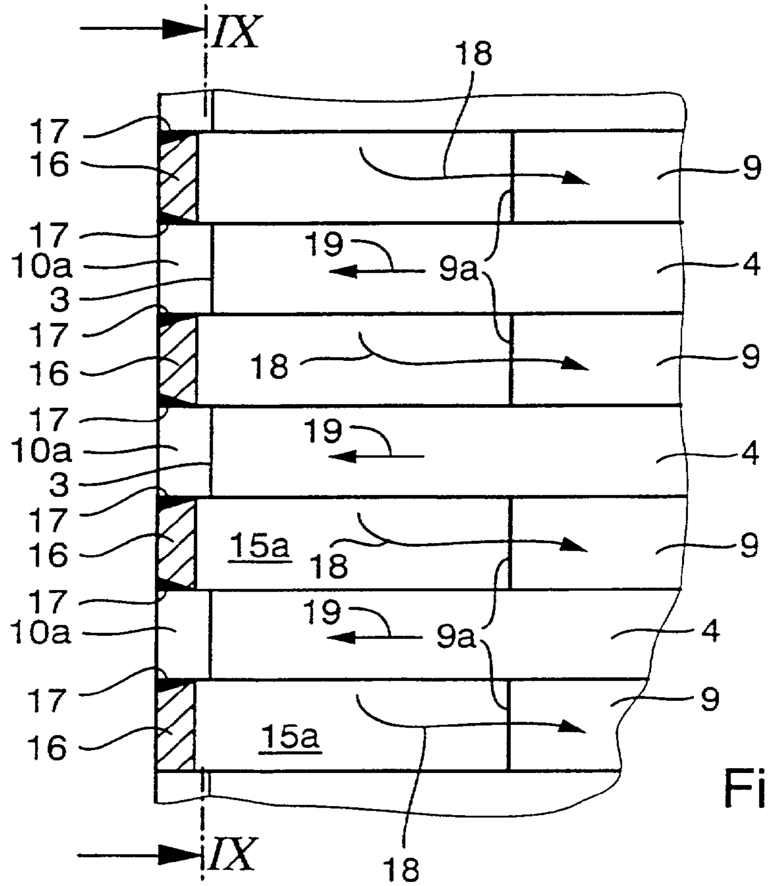


Fig. 7


IX
18
17
16
9
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10a
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9a
4
3
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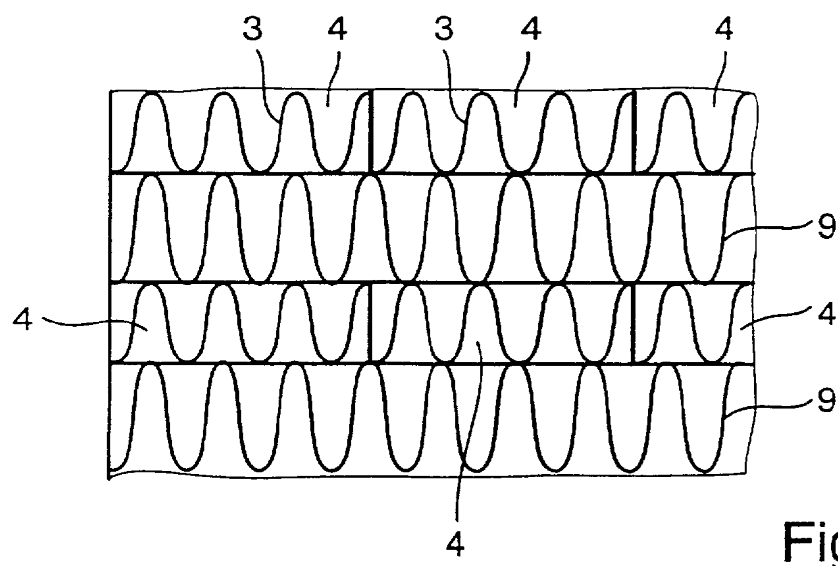
15a
IX
18

Fig. 8


3
4
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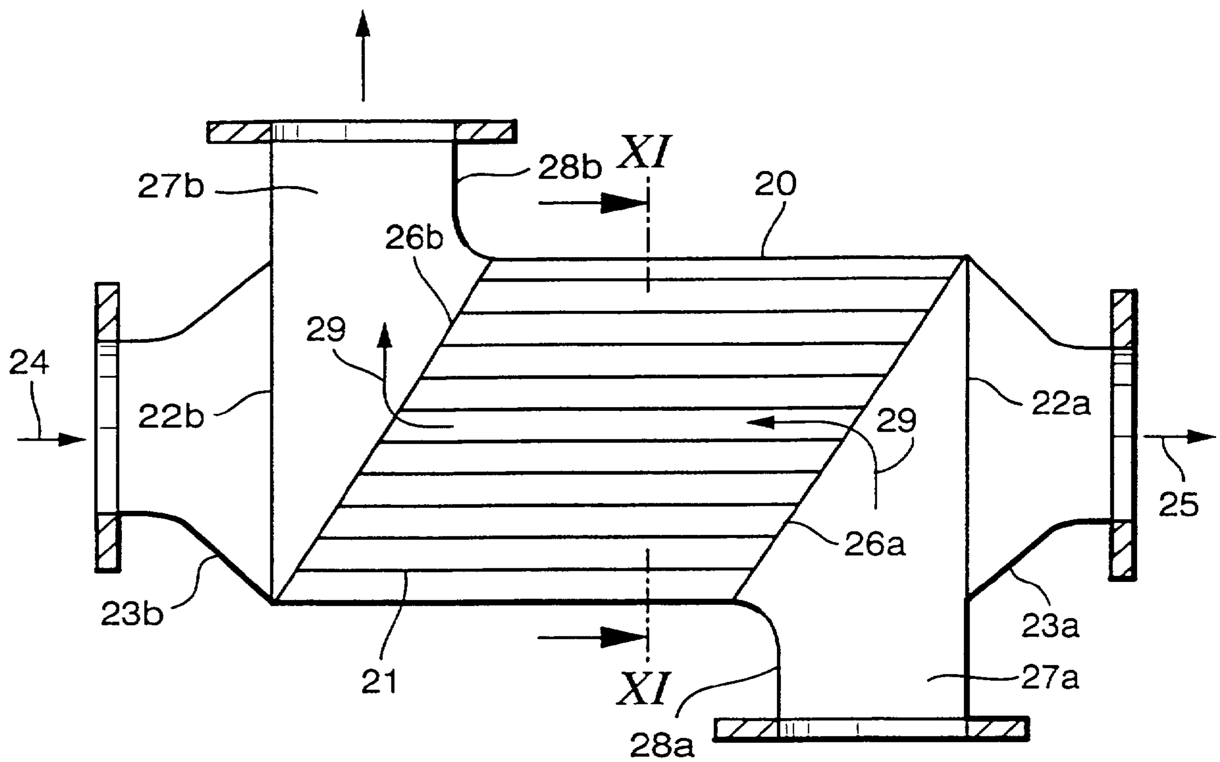
4

Fig. 9


XI
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27b
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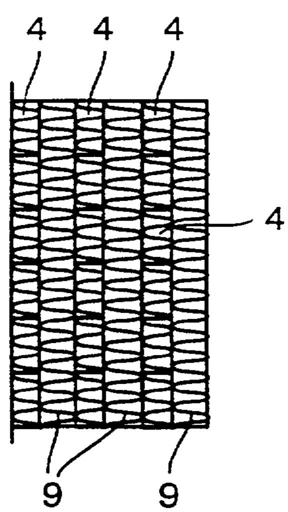
26b
29
24
22b
22a
29
25
26a
23b
23a
21
XI
27a
28a

Fig. 10


4
4
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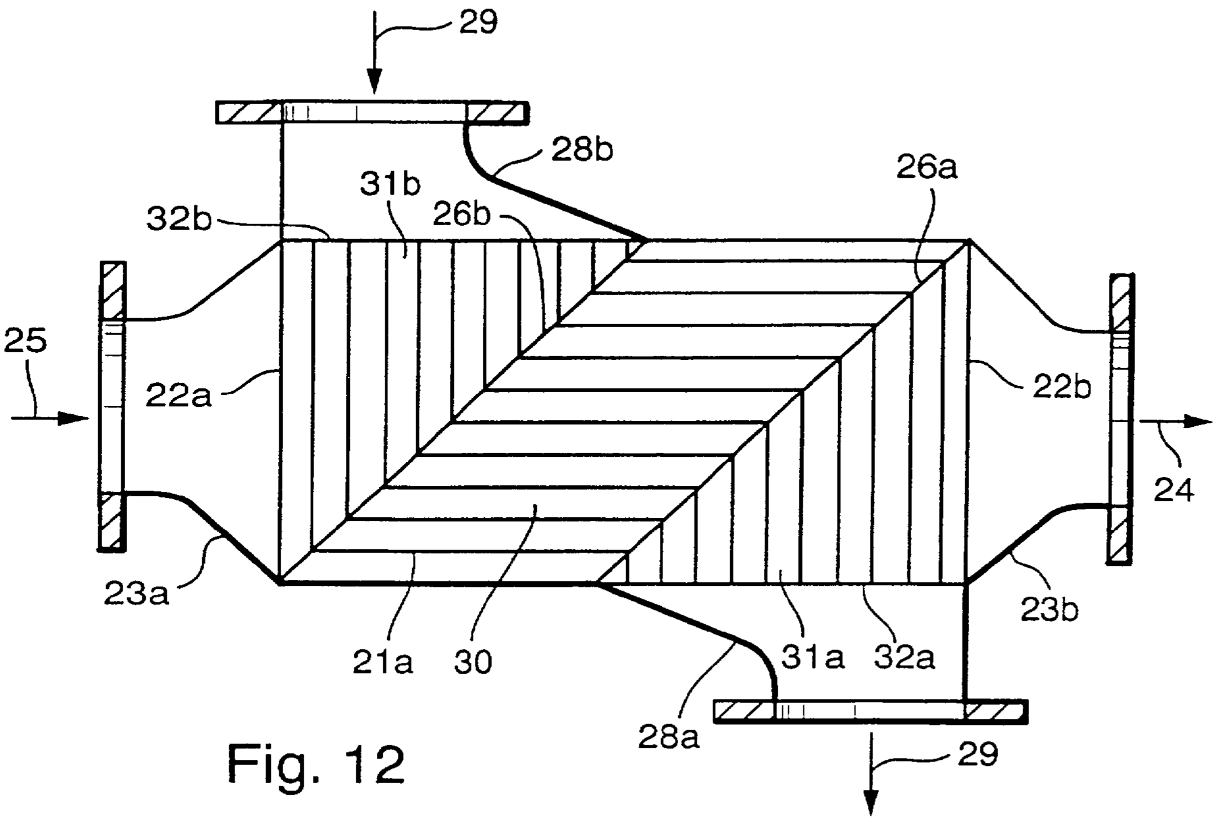
9
9

Fig. 11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26a
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32b
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22a
22b
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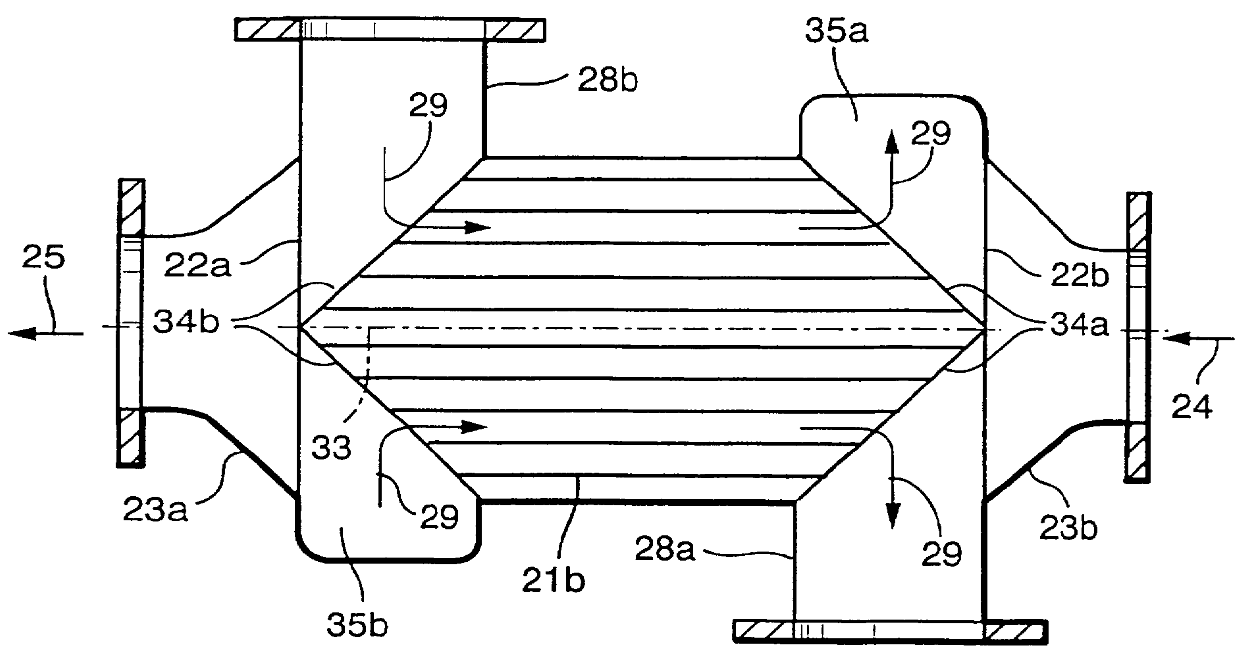
23a
23b
21a
30
31a
32a
28a
29

Fig. 12


35a
28b
29
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22a
22b
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34b
34a
24
33
23a
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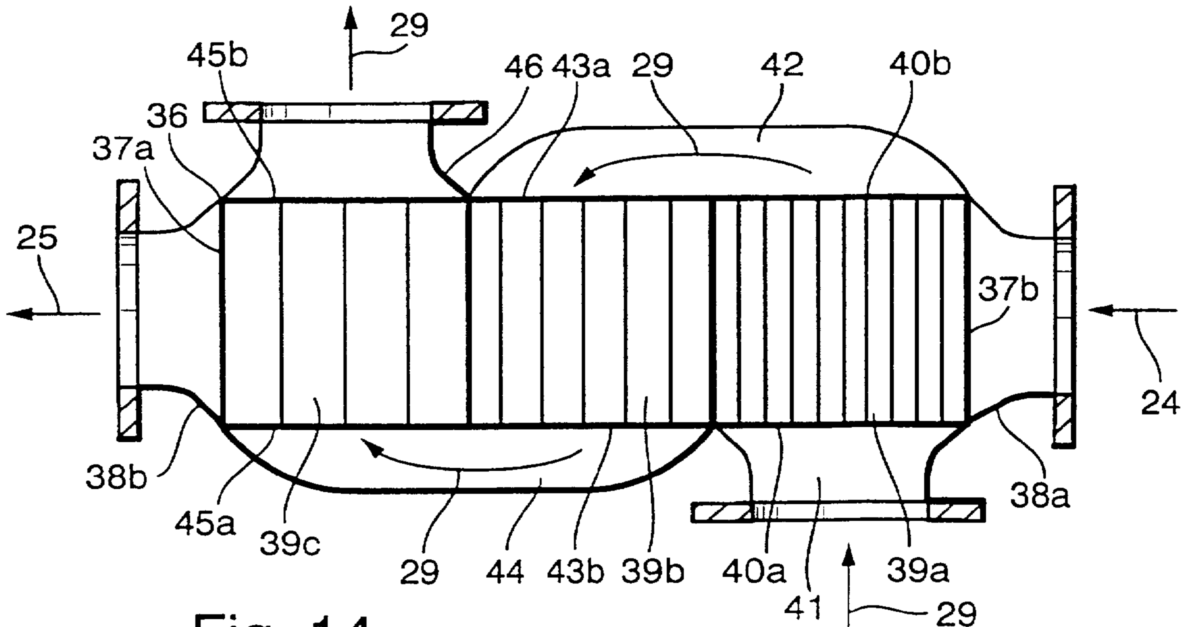
23b
28a
29
21b
35b

Fig. 13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45b
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43a
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42
40b
36
37a
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37b
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38b
38a
45a
39c
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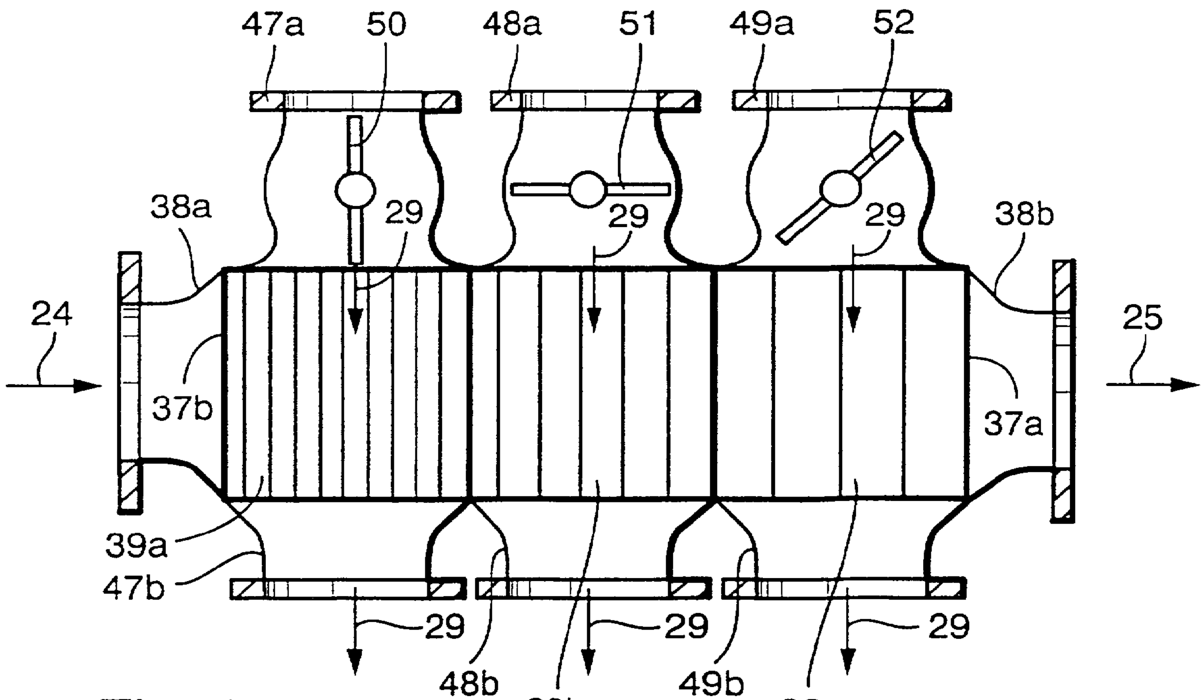
44
43b
39b
40a
39a
41
29

Fig. 14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49a
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38a
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38b
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37a
39a
47b
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29
29
48b
39b
49b
39c

Fig. 15

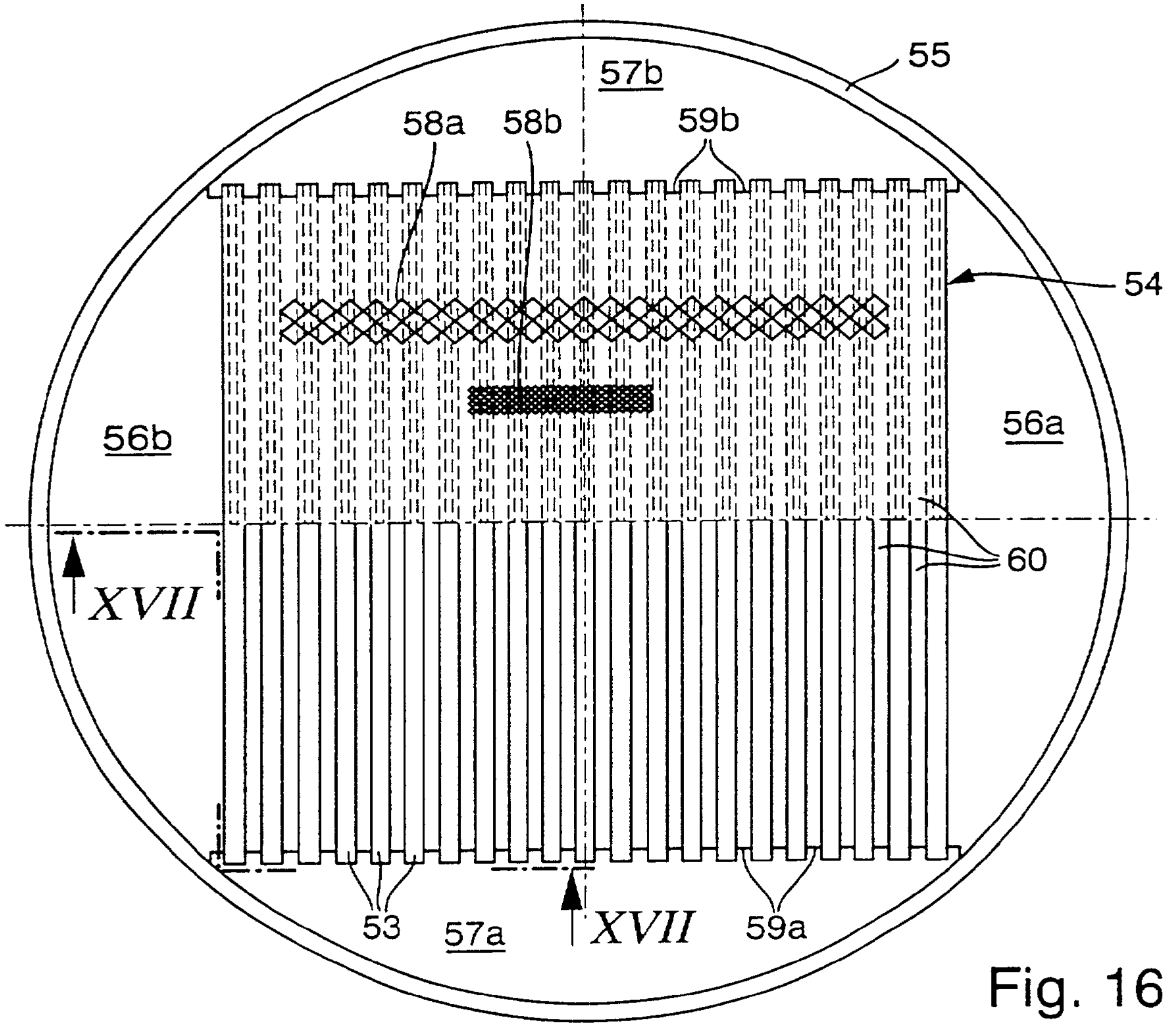

Fig. 16

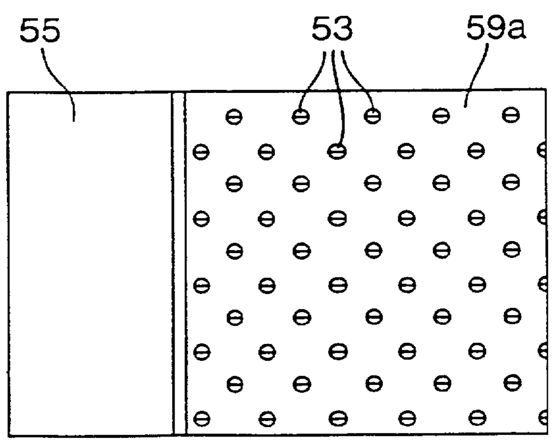

Fig. 17

Reactor with a heat exchanger structure

The invention relates to a reactor for catalytically converting a flow of a fluid operating substance, having a heat exchanger structure designed as a bank of tubes with at least a first and, in heat contact therewith, a second heat-exchanger chamber, the first thereof containing a reaction chamber coated with a conversion catalyst for converting the flow of operating substance delivered thereto. Depending on requirements, as a result of the heat exchanger structure, heat can be applied to or dissipated from the reaction chamber, being one of at least two heat exchanger chambers disposed in heat contact with one another while being fluidly separated, an appropriate heat exchange medium being circulated through the other heat exchanger chamber. Accordingly, defined temperature conditions can be set in the reaction chamber in order to conduct endothermic, exothermic or autothermic reactions.

Particularly for heterogeneously catalysed reactions, there is a need for reactors which will provide as large a contact surface area as possible and guarantee good temperature control between the flow of operating substance to be converted and the conversion catalyst, so that the conversion reaction can be operated to a high degree of efficiency and selectivity. A known type of reactor used for the purpose of this application is the fixed bed reactor, the reaction chamber of which is filled with catalytic pellets. However, these reactors require a relatively large construction volume and have the disadvantage of peripheral skirting caused by bypass flows in the granulate and between the pellets and the reactor walls. Furthermore, the heat conductivity of pellet fillings is relatively low causing an uneven temperature profile in the reaction chamber, which can reduce the efficiency and selectivity of the conversion reaction. The reactors needed for certain specific applications, e.g. for running reforming reactions to generate hydrogen in mobile applications, are required to have a high heat flow density in the direction of flow, if necessary being variable, to be able to guarantee that the process can be run optimally and the reactor operated with a sufficiently dynamic operating behaviour.

In addition to packing with conversion catalyst in the form of pellets, a known approach is to coat the interior wall of the reaction chamber with a conversion catalyst. Publication DE 196 53 991 discloses a reactor of this type, specifically designed for implementing endothermic catalytic reactions, the heat-exchanger structure of this reactor consisting of a monolithic arrangement of heating and reaction passages running parallel with one another. Whilst the interior wall surfaces of the reaction passages are coated with the conversion catalyst, the interior

wall surfaces of the heating passages are coated with a combustion catalyst to enable a catalytic combustion process to be induced there, by means of which the reaction passages forming the reaction chamber are heated. The hot combustion gas resulting from the combustion process can be circulated through the heating passages in counter-flow or parallel flow with the flow of operating substance circulating through the reaction passages. Providing the catalyst in the form of a coating on the interior wall avoids the problems inherent in pellets, such as peripheral skirting, low heat conductivity, uneven temperature distribution and large construction volume, but the specific reaction surface area, i.e. the effective catalytic surface area, is relatively limited because the conversion catalyst is present only on the interior wall of the reaction chamber.

The present invention seeks to provide a reactor of the type outlined above, which is of a relatively small construction volume and weight for a given operating capacity and has a high specific reaction surface area whilst avoiding the effects of peripheral skirting.

According to the present invention there is provided a reactor for catalytically converting a flow of operating substance having

- a heat exchanger structure in the form of a bank of tubes with at least a first and, in heat contact therewith, a second heat exchanger chamber, the first of which chambers contains a reaction chamber with a conversion catalyst for converting the flow of operating substance delivered thereto, wherein
- a heat-conductive, corrugated rib structure coated with the conversion catalyst is located in the reaction chamber.

With this reactor, a heat-conductive, corrugated rib structure is inserted in the reaction chamber, which is coated with the conversion catalyst. The conversion catalyst may be applied by coating the corrugated rib surface with a wash-coat or by subjecting the corrugated rib surface to a surface treatment to produce micro-pores and then incorporating or depositing the conversion catalyst in or on the corrugated rib surface enlarged in this manner.

Because the corrugated, ribbed surface in the reaction chamber coated in this way can be significantly larger than the surface area of the walls externally bounding the reaction chamber, a correspondingly larger specific reaction surface area can be provided than is the case in a reactor in which merely the walls externally bounding the reaction chamber are coated with the conversion catalyst. At the same time, the difficulties mentioned above in connection with

catalyst pellets, such as peripheral skirting and uneven temperature distribution, are avoided if using the reactor proposed by the invention. Furthermore, providing the conversion catalyst as a coating on corrugated ribs inserted in the reaction chamber provides favourable prerequisites for obtaining the necessary density of heat flow depending on load and spatially defined in order to ensure the most efficient dynamic operating behaviour possible. The reactor design proposed by the invention is strong enough to withstand mechanical stress and is not susceptible to friction and ageing of the conversion catalyst. The reactor is suitable for implementing both exothermic and endothermic or autothermic chemical reactions, for which purpose an appropriate heat exchange medium can be circulated through the other heat exchanger chamber in heat contact with the reaction chamber, either as a heating or cooling medium.

In another embodiment of the reactor, the reaction chamber is formed by the interior volume of a bank of flat tubes, in which corrugated ribs coated with the conversion catalyst are inserted. The bank of flat tubes may be a variable number of flat tubes, e.g. with a rectangular cross section, through which a flow can be circulated in parallel and which together form the reaction chamber.

In another embodiment of the invention, the flat tubes may each be made up of two tube half-shells, which are pre-fabricated to have a cross section which widens towards the exterior. As a result of this cross section shaped so that it is wider towards the side regions, the two tube half-shells can be mounted so that they are mechanically pre-stressed against a corrugated rib, coated with the catalytic material at least at the rib flanks, being designed to contain the latter, and then joined to one another at the edge faces in order to form the flat tube. Accordingly, the flat tube lies tightly biased against the individual arches of the corrugated rib disposed inside it, producing a good heat contact between corrugated rib and flat tube wall.

In another embodiment of the invention, the heat exchanger structure comprises a flat tube/rib block, in which the plurality of flat tubes forming the reaction chamber in which the corrugated ribs coated with the catalytic material are inserted, are spaced at a distance apart and have heat-conductive corrugated ribs disposed between them on the outside of the flat tubes. As a result, the intermediate spaces between the flat tubes of the flat tube/rib blocks form a heat exchanger chamber which is in heat contact with the reaction chamber formed by the interiors of the flat tubes and in which heat-conductive corrugated ribs are also mounted in order to improve the heat-transfer capacity. In another embodiment of this type of reactor, the flat tubes are widened to such a large degree at the end face that the flat tube ends bridge the intermediate

spaces between flat tubes and lie one against the other to form a solid join, which may be provided with an appropriate connecting structure to direct the flow of operating fluid used in and direct the flow of resultant product out.

In another embodiment of the invention, the flat tube/rib block is mounted in a heat exchanger housing and the heat exchanger chamber on the outside of the flat tubes has flow passages running in the longitudinal direction of its associated corrugated ribs. Depending on the layout of these corrugated ribs on the outside of the flat tubes relative to the corrugated ribs coated with the conversion catalyst inside the flat tubes, the heat exchange medium can be directed as desired, e.g. in parallel flow or alternatively in cross-flow to the flow of operating fluid directed through the reaction chamber. Appropriate connecting structures are provided on the heat exchanger housing to direct the heat exchange medium on the one hand and the flow of operating medium on the other in and out. In another embodiment of this reactor type, the heat exchanger chamber through which the heat exchange medium is circulated can be divided into several part-compartments by designing its connecting structure accordingly, so that the heat exchange medium can be circulated in parallel or in series.

In another embodiment of the invention, the reaction chamber comprises a stack of corrugated ribs coated with the conversion catalyst, lying one on top of the other at a predeterminable cross-over angle and the resultant stack of corrugated ribs has a bank of parallel heat exchange tubes spaced apart from one another passing through it, the tube interior in this embodiment forming the heat exchanger chamber through which the heat exchange medium is circulated.

Advantageous embodiments of the invention will be described below, by way of example, with reference to the drawings, in which:

Fig. 1 is a side view of a corrugated rib-blank prior to coating with a conversion catalyst,

Fig. 2 is a side view of the corrugated rib illustrated in Fig. 1 after being coated with the conversion catalyst,

Fig. 3 is a view in cross section of a flat tube made up of two tube half-shells which will contain the coated corrugated rib illustrated in Fig. 2, prior to assembling the tube half-shells,

Fig. 4 shows the assembled flat tube illustrated in Fig. 3 after fitting the two tube half-shells together,

Fig. 5 is a view in cross section of a flat tube/rib block, made up of flat tubes as illustrated in Fig. 4 and intermediately inserted corrugated ribs,

Fig. 6 is a side view of an assembled flat tube/rib block of the type illustrated in Fig. 5,

Fig. 7 is an end view onto the flat tube/rib block of Fig. 6,

Fig. 8 is a schematic cut-away side view of a flat tube/rib block which has been modified compared with that of Fig. 6, having a tube plate instead of widened flat tube ends,

Fig. 9 is a cut-away view of a section along the line IX-IX of Fig. 8,

Fig. 10 is a longitudinal section of a reactor with a flat tube/rib block, designed for parallel flow, enclosed in a housing,

Fig. 11 is a half section along the line XI-XI of Fig. 10,

Fig. 12 is a longitudinal section of a reactor with the flat tube/rib block mounted in the housing, the flow being directed along a Z-shaped course,

Fig. 13 is a longitudinal section of a reactor with the flat tube/rib block mounted in the housing, the flow being directed along a double-Z-shaped course,

Fig. 14 is a longitudinal section of a reactor with the flat tube/rib block mounted in a housing, with a heat exchanger chamber divided into three parts in series by means of an appropriate connecting structure,

Fig. 15 is a longitudinal section of a reactor with the flat tube/rib block mounted in a housing, with a heat exchanger chamber divided into three parallel parts by an appropriate connecting structure,

Fig. 16 is a schematic cross section of a reactor with a heat exchanger structure, comprising a stack of corrugated ribs and a bank of heat exchange tubes inserted through them, mounted in a housing and

Fig. 17 is a half section of the reactor along the line XVII-XVII of Fig. 16.

Fig. 1 illustrates a corrugated rib blank 1 in the form used to made a catalytically coated corrugated rib. Fig. 2 shows a corrugated rib 3 made from the blank 1 with a coating 2 of a conversion catalyst. In order to apply the layer of catalyst 2, the surface of the corrugated rib blank 1 may firstly be provided with a wash-coat to enlarge the surface area or may be subjected to a surface treatment to create micro-pores. The corrugated rib blank 1 is typically a metal strip 0.02 mm to 0.3 mm thick made from aluminium, stainless steel, copper or similar.

In order to produce a plurality of very fine micro-pores, a corrugated rib blank may be subjected to a process of anodic oxidation in sulphuric acid or another electrolyte, such as carbonic acid, for example, producing a metal oxide surface layer containing micro-pores, see the dissertation by D. Scholl, University of Karlsruhe, 1989. The micro-pores, of a blind bore shape, are disposed exclusively in the oxide layer and do not therefore continue down into the metal, thereby simultaneously providing a relatively anti-corrosive surface. The nature of the pore system in terms of pore density, pore diameter and pore length can be controlled by a series of parameters such as the type of electrolyte, voltage and timing applied during the anode oxidation process and therefore duly adapted to the desired requirements. Typical sizes are 10μm to 300 μm for the pore length and 10 nm to 100 nm for the pore diameter. The micro-pores formed are then doped with the conversion catalyst, for example by dipping in a solution followed by firing or by an appropriate CVD or PVD process. By preference, appropriate steps are taken to ensure that the outer face 3a of the arches of the corrugated rib held flat are not perceptibly coated with the catalytic material, for example by masking the corrugated rib with a non-removable coating on both sides at the outer ends during the coating process or by scraping off the wash-coat after the coating process.

Figs. 3 and 4 illustrate the procedure for manufacturing a flat tube 4 which may be used for the purpose of the invention, having a coated corrugated rib 3 such as that illustrated in Fig. 2 inserted inside it. First of all, two matching tube half-shells 4a, 4b are pre-fabricated, the shape of which in cross section can be seen from Fig. 3 at the pre-fabricated stage. As may be seen, the tube half-shells 4a, 4b extend from their turned down side edges 5 in a slightly V-shaped incline towards their centre region 6. As a result, once they have been laid around either side of the corrugated rib 3 they are to enclose and their down-turned side edges 5 have been pushed together in abutment where they are permanently joined to one another by a weld seam 7, they lie mechanically prestressed against the corrugated rib arch 3a. As a result, the corrugated rib 3 and the flat tube wall are in a close, forced and heat conductive contact, which provides an effective heat contact between the heat-conductive corrugated rib 3 and the tube wall of the flat tube 4, and hence a good transport of heat between the interior 8 of the flat tube 4 and the area external to it. The two half-shells 4a, 4b are preferably made from stainless steel and may be made endlessly by folding a strip of material which will have a typical thickness of between 0.05 mm and 0.4 mm. In order to provide as large a heat exchange surface area per unit volume as possible to obtain a compact reactor structure, a relatively fine rib structure is chosen for the corrugated rib 3, typically with rib densities of between 15Ri/dm and 150Ri/dm. The finished flat tube 4 is preferably between 10 mm and 50 mm wide and between 1 mm and 6 mm high.

Fig. 5 is a cut-away illustration showing one possible structure of a reactor using the flat tube 4 illustrated in Fig. 4. Specifically, the flat tube/rib block preferably contains several flat tubes 4, with a corrugated rib 3 coated with the conversion catalyst disposed inside, in a one- or two-dimensional layout, the flat tubes being spaced apart from one another in a direction of height H by mounting second corrugated ribs 9 on the external face of the flat tubes one after the other. Fig. 6 illustrates a flat tube/rib block of this type with ten layers of flat tubing, for example.

As may also be seen from Fig. 6, the flat tubes 4 extend out at their ends 10 beyond the intermediately inserted corrugated ribs 9 on the outside of the flat tubes and are widened to such a degree that the ends of each two adjacent flat tubes 4 at the same side lie in contact with one another and therefore close off the respective space between the flat tubes at the outer end. The walls of the flat tube ends 10 lying one against the other are joined to one another by a soldered or welded joint 12 so as to be fluid-tight.

Fig. 7 shows the flat tube/rib block of Fig. 6 in an end view along an arrow S_1 shown in Fig. 6 for an embodiment having in a block depth direction T two rows 11a, 11b of flat tubes 4

adjoining one another at the side. To produce this arrangement, as may be seen from Fig. 7, two flat tubes in each tube block layer are laid adjacent to one another and in contact and may be permanently secured to one another by a soldered or welded joint 13 along their touching side walls if necessary, whilst each of two flat tubes 4 of a respective row of flat tubes 11a, 11b lying one underneath the other are secured to one another at their end regions 10 by soldered or welded joints 12 at the end face. As a result, the flat tube/rib block forms, at its two side end faces, an opening structure across its entire surface for the medium to be circulated through the flat tube interiors 8, as may be seen from Fig. 7, without the need for distributor tube plates in which the flat tube ends would otherwise be inserted.

Specifically, the flow-parallel flat tube interiors 8 of the flat tube/rib block illustrated in Figs. 5 to 7 form a reaction chamber of a reactor for catalytically converting an operating substance flow fed to them, the catalytic material for the conversion with which the corrugated ribs 3 inside the flat tubes are coated being selected so that it catalyses the desired conversion reaction. The intermediate spaces between the flat tubes in which the corrugated ribs 9 are mounted on the outside of the flat tubes then form a second heat exchanger chamber in heat contact with the reaction chamber 8 serving as a first heat exchanger chamber, through which an appropriate heat exchange medium can be circulated. Depending on the application and depending on whether an exothermic, endothermic or autothermic conversion reaction is to take place, this may be a heating or cooling medium. To this end, the corrugated ribs 9 on the outside of the flat tubes are also designed to be heat-conductive and may be made from the same material as the corrugated ribs 3 on the inside of the flat tubes. The heat exchange medium can be introduced into the intermediate spaces between the flat tubes via side gaps 15 formed between the flat tube ends 10 lying against one another on the one hand and the recessed ends 9a of the corrugated ribs 9 on the outside of the flat tubes, on the other hand, where it flows in parallel flow, more specifically depending on the chosen inflow and outflow system, in co-current or counter-flow, with the operating flow, which is circulated through the flat tube interiors 8.

If the heat exchange medium is to be a heating medium, the corrugated ribs 9 on the outside of the flat tubes may be provided with a coating 14 of a combustion catalyst, similarly to the corrugated ribs 3 on the inside of the flat tubes, as illustrated in Fig. 5, which will cause a catalytic combustion of a flow of fuel fed in. The coating 14 of combustion catalyst may be applied to a corrugated rib-blank in the same way as the corrugated ribs 3 are coated with the conversion catalyst as described above.

The flat tube/rib block can be put together firstly by loosely placing the individual flat tubes 4 and the corrugated ribs 9, lying outside and between the flat tubes, together and then bracing them as a pack in a housing and/or soldering them together in a common soldering process. If the corrugated rib arches are left uncoated, the corrugated ribs 9 can be more easily soldered to the flat tubes 4.

In the flat tube/rib block illustrated in Figs. 5 to 7, the corrugated ribs 14 on the outside of the flat tubes run with their longitudinal axis parallel with the longitudinal axis of the corrugated ribs 3 on the inside of the flat tubes. If, as illustrated, they are placed against the flat tube external wall with the corrugated rib arches flush with the arches of the corrugated ribs 3 on the inside of the flat tubes, the structure produced is particularly strong and allows the use of very wide flat tubes without the need to use reinforcing inserts. Furthermore, because any number of flat tubes 4 can be assembled with one another in a modular two-dimensional block arrangement, the flat tube/rib block can be made to any desired dimensions of height and depth. For applications in which no external support is provided by corrugated ribs on the outside of the flat tubes, it is preferable to use slimmer flat tubes to enable them to withstand compression stress as required. As an alternative to the longitudinal ribbing illustrated, i.e. in which the corrugated ribs on the outside of the flat tubes are mounted with their longitudinal direction parallel with the longitudinal direction of the flat tubes, it is also possible to make the flat tube/rib block with cross-ribbing, in which case the corrugated ribs on the outside of the flat tubes are mounted in the intermediate spaces between the flat tubes with their longitudinal direction shifted, preferably in a perpendicular longitudinal direction. This being the case, the flow passages of the heat exchange medium-heat exchanger chamber defined by the corrugated ribs on the outside of the flat tubes and the external walls of the flat tubes will extend perpendicular to the reaction chamber flow passages, each of which is defined by the corrugated rib on the inside of the flat tube and the internal wall of the flat tube, i.e. the heat exchange medium will be circulated in cross-flow to the operating substance flow used.

Figs. 8 and 9 are sections showing a different embodiment of the flat tube/rib block illustrated in Figs. 5 to 7, in which the flat tube ends are not widened but are inserted in a tube plate so as to be fluid-tight. To this end, a tube plate 16 of the type illustrated in Fig. 4 is welded between the ends 10a of flat tubes 4 at the same end by appropriate weld joints 17 so that the corrugated ribs 3 on the inside of the flat tubes terminate short of the level of the tube plate and the tube plate 16 closes off the intermediate spaces between the flat tubes at the side. The heat

exchange medium 18 is fed through gaps 15a between the tube plate 16 and the ends 9a of the corrugated ribs 9 outside the flat tubes set back from it, whilst the operating flow 19 is fed through the reaction chamber into and out of the flat tube openings at the end faces. In the section illustrated in Fig. 9, two layers of flat tubes each with three flat tubes 4 lying in contact with one another are illustrated as an example of a cut-out section from the entire flat tube/rib block, in which an end-to-end corrugated rib 9 on the outside of the flat tubes is mounted between every two flat tube layers to provide longitudinal ribbing. Alternatively, the corrugated ribs on the outside of the flat tubes can also be provided as cross-ribbing in this embodiment, with the longitudinal axis of the corrugated ribs running perpendicular to the longitudinal axis of the flat tube.

Figs. 10 to 15 illustrate various possible reactor structures, in each of which a flat tube/rib block is mounted in a housing to provide a heat exchanger structure, the appropriate connecting structures being provided for directing the operating substance flow used and the resultant product flow in and out of the flat tube interiors and for directing the heat exchange medium in and out of the intermediate spaces between the flat tubes, which are also ribbed.

The reactor illustrated in Figs. 10 and 11 is one with a partially ribbed structure with a Z-shaped circulation design. To be more specific, the flat tubes 4 of the flat tube/rib block 21 mounted in a housing extend between two oppositely lying, parallel tube plates 22a, 22b, each of which has its own housing connection 23a, 23b at the end face in order to direct the flow of operating substance 24 to be converted into the reaction chamber formed by the flat tube interiors and to direct the generated product flow 25 out at the opposite end. The corrugated ribs 9 on the outside of the flat tubes, in the intermediate spaces between flat tubes, terminate along two closure lines 26a, 26b running at an incline to the tube plates 22a, 22b leaving a distance to the tube plates 22a, 22b and forming respectively a wedge-shaped, non-ribbed inflow region 27a and outflow region 27b, each of which is provided with a radial housing connection 28a, 28b. As a result, the heat exchange medium 29 is fed through the housing 20 in a Z-shaped pattern so that it circulates in the effective heat exchange region of the flat tube/rib block 21 in counter-flow with the operating substance to be converted. In a half cross section, Fig. 11 shows the flat tube/rib block illustrated in Fig. 10 in an example in which the illustrated block half has three rows each with six flat tubes 4 lying in contact one underneath the other, alternating with three end-to-end corrugated ribs 9 outside the flat tubes providing longitudinal ribbing.

The reactor illustrated in Fig. 12 essentially corresponds to that illustrated in Figs. 10 and

11, and, for the sake of simplicity, the same reference numbers are used to denote elements performing the same function and details of these can be obtained from the description given above in conjunction with Figs. 10 and 11. Unlike the reactor illustrated in Figs. 10 and 11, the ribbing of the flat tube/rib block 21a on the outside of the flat tubes starting from a ribbing centre region 30, with the longitudinal axis of the corrugated ribs extending parallel with the flat tube longitudinal axis, does not terminate on a level with the two inclined lines 26a, 26b but continues on, having a respective mitre joint with the ribbing centre region 30, into a ribbing 31a, 31b, each of the end faces 32a, 32b of which has a radial housing connection 28a, 28b. As a result, the heat exchange medium 29 in the two ribbing part-regions 32a, 32b at the ends is circulated in cross flow and in the ribbing centre region 30 in counter-flow to the flow of operating substance 24 to be converted and the product flow 25 passing the reaction chamber.

Another alternative with regard to circulating the heat exchange medium 29 is illustrated in the reactor of Fig. 13, in which the same functional elements are again shown by the same reference numbers as those used for the reactor of Fig. 10 and 11 and reference should be made to the relevant description for details of these. In the reactor illustrated in Fig. 13, a flat tube/rib block 21b is used, in which the longitudinal ribbing on the outside of the flat tubes terminates symmetrically with a longitudinal central plane of the housing along a rib end-surface 34a, 34b extending at an incline relative to the tube plate 22a, 22b, each of which is provided with a circumferential inflow and outflow ring 35a, 35b as a connecting structure, into which the associated radial housing connection 28a, 28b opens. The result is a double-Z-shaped circulation through the flat tube/rib block 21b by the heat exchange medium 29 in counter-flow with the operating substance 24 to be converted and the product flow 25 circulating through the flat tube interiors.

In the reactor embodiment illustrated in Fig. 14, a flat tube/rib block 37 is mounted in a housing 36, the flat tubes of which again extend in a straight line between two oppositely lying tube plates 37a, 37b, being provided respectively with a housing connection 38a, 38b for feeding in the flow of operating medium 24 to be converted and for feeding away the resultant product flow 25. The ribbing of the flat tube/rib block 37 on the outside of the flat tubes is designed as cross-ribbing, i.e. the corrugated ribs on the outside of the flat tubes run with their longitudinal axis perpendicular to the longitudinal axis of the flat tubes. Due to a specially provided connecting structure, the heat exchange medium-heat exchanger chamber of the flat tube/rib block containing the cross-ribbing is divided into three parts 39a, 39b, 39c, in series one after the other.

The inlet end 40a of the upstream part 39a is provided with a radial inlet connector 41, whilst the outlet end 40b of the upstream part 39a of the heat exchange medium-heat exchanger chamber at the opposite end of the housing is in fluid contact with the inlet side 43a of the centre part 39b via a first housing-end baffle 42. The outlet end 43b of the centre part 39b is connected to the inlet end 45a of the downstream part 39c of the heat exchange medium-heat exchanger chamber by means of a second housing-end baffle 44. The outlet end 45b thereof adjoins a radial outlet connector 46.

Accordingly, in the reactor illustrated in Fig. 14, the heat exchange medium 29 is directed in cross-flow to the flow of operating substance 24 and resultant product flow 25 and, after entering the inlet connector 49, circulates through the upstream part 39a of the associated cross-ribbed heat exchanger chamber, from where it is deflected into the central part 39b, flows through it and is then deflected in the downstream part 39c and, after flowing through it, is fed out of the reactor housing via the outlet connector 46.

Different ribbing may be selected for the three parts 39a, 39b, 39c of the heat exchange medium-heat exchanger chamber. For example, part 39a at the inlet end could contain corrugated ribs with a coating of combustion catalyst, whilst the corrugated ribs in the central part 39b could operate merely as supporting and heat-conductive ribs without any coating and the downstream part 39c might have no ribs at all. This would be a simple means of producing a drop in temperature along the flow path through the reaction chamber.

Fig. 15 illustrates a different embodiment of the reactor illustrated in Fig. 14, elements performing the same function again being denoted by the same reference numbers. The reactor illustrated in Fig. 15 differs from that illustrated in Fig. 14 due to the fact that the three parts 39a, 39b, 39c of the heat exchange medium-heat exchanger chamber are not disposed in series but are connected in parallel. To this end, each of the three parts 39a, 39b, 39c has its own connection system with a radial inlet connector 47a, 48a, 49a and a radial outlet connector 47b, 48b, 49b lying opposite. Disposed in each of the three radial inlet connectors 47a, 48a, 49a is a controllable throttle valve 50, 51, 52 by means of which the through-flows of heat exchange medium 29 circulating through each of the three parts 39a, 39b, 39c of the associated heat exchanger chamber can be individually regulated. As a result, very variable temperature distributions can be set in the flow path along the reaction chamber formed by the flat tube interiors, for example a higher temperature in the region of the inlet end of the reaction chamber and a lower temperature in the region of the outlet end of the reaction chamber.

Again with the reactor illustrated in Fig. 15, different ribbing may be selected for the three parts 39a, 39b, 39c of the heat exchange medium-heat exchanger chamber in order to obtain a desired temperature profile, as explained above with reference to the reactor illustrated in Fig. 14.

Clearly, in the different embodiments of reactors described above, it would also be possible to select reverse flow directions for the heat exchange medium and operating substance to be converted instead of those illustrated. Furthermore, the reactors may also be used with reversed roles of heat exchange medium and operating substance to be converted, i.e. in this case, the corrugated ribs on the outside of the flat tubes of the flat tube/rib block would be coated with the conversion catalyst whilst the corrugated ribs inside the flat tubes would be left uncoated or would be coated with a combustion catalyst, for example. This being the case, the reaction chamber would be formed in the intermediate spaces between tubes whilst the flat tube interiors would constitute the heat exchange medium-heat exchanger chamber.

Figs. 16 and 17 illustrate another type of reactor proposed by the invention, in which a stack of corrugated ribs coated with a conversion catalyst layered one above the other crossways at a crossover angle of 90° having heat exchange tubes 53 passing through them is mounted in a cylindrical housing 55 as a heat exchanger structure 54 which is more or less square-shaped in cross section. The four segment-shaped volumes left between the die-shaped or cuboid heat exchanger structure 54 and the cylindrical housing 55 form, firstly, two oppositely lying connecting chambers 56a, 56b for the operating substance flow to be converted as it flows into the flow passages formed by the alternating corrugated ribs lying crosswise one above the other, and secondly, two oppositely lying connecting chambers 57a, 57b for a heat exchange medium circulated through the heat exchanger-round tubes 53.

In order to make the heat exchanger structure 54, depending on requirements, corrugated ribs 58a with a lower rib density or corrugated ribs 58b with a higher rib density may be used, as schematically illustrated in an insert in Fig. 16. If necessary, the stack of corrugated ribs may also be constructed of corrugated ribs with a different rib density. The round tubes 53 are inserted through circular orifices provided in the corrugated ribs as a two-dimensional bank of tubes spaced apart, as may be seen from Fig. 17, and then secured to the corrugated rib stack by mechanical or hydraulic widening. At the end face, the heat exchange tubes 53 are inserted respectively in a tube plate 59a, 59b, the tube plates 59a, 59b sealing off the reaction chamber formed by the corrugated rib passages 60 between heat exchange tubes 53 from the two heat exchange medium-connecting chambers 57a, 57b.

In this design of reactor, the heat exchange medium fed through the heat exchange tubes 53 within the heat exchanger structure 54 flows in cross-flow to the operating substance flow through the stack of corrugated ribs lying crosswise one above the other and coated with conversion catalyst, on which the desired chemical conversion reaction takes place. The conversion heat generated or required is transferred to the heat exchange medium via the round tubes or is imparted thereby. In order to produce a good transfer of heat, the round tubes 53 are of a relatively small diameter, in the order of between 1 mm and 5 mm. In addition, the round tubes 53 are arranged at a relatively small transverse and longitudinal distance of 2 mm to 10 mm. The corrugated ribs are again typically 0.02 mm to 0.3 mm thick with a rib density of 50Ri/dm to 150Ri/dm.From the detailed description of embodiments given above, it is clear that the reactor proposed by the invention can be made to a relatively small structure and will have a high specific reaction surface area with an appropriate conversion catalyst system, exhibiting no peripheral skirting effects and no bypass effects, producing constant diffusion paths and residence times in the reaction chamber for the flow of operating substance. This results in a very selective and efficient reaction, a uniform temperature distribution and, if required, a good and locally defined input of heat into the system. The reactor exhibits good dynamic operating behaviour and high mechanical strength. The conversion catalyst is disposed friction-free in the reaction chamber. If only welded joints rather than soldered joints are used for fixing purposes, it will be possible to avoid the risk of introducing any undesirable materials which might have a catalytic effect, as would be the case with solder. The highly modular variety of structures in which the reactor can be assembled means that it can be made to produce the most varied of flow circulation patterns, such as counter-flow, co-current-flow and cross-flow and combinations thereof, and the most varied of conversion outputs and in different reactor stages at different reaction temperatures. The reactor proposed by the invention may be used for any catalytic chemical process, including in particular hydrogen-generating reforming reactions in stationary and mobile applications, e.g. for steam reforming methanol, and as an oxidation stage downstream of a reformer to remove carbon monoxide from the untreated gas.

Claims

1. A reactor for catalytically converting a flow of operating substance having

- a heat exchanger structure in the form of a bank of tubes with at least a first and, in heat contact therewith, a second heat exchanger chamber, the first of which chambers contains a reaction chamber with a conversion catalyst for converting the flow of operating substance delivered thereto, wherein
- a heat-conductive, corrugated rib structure coated with the conversion catalyst is located in the reaction chamber.

2. A reactor as claimed in claim 1, wherein the reaction chamber is formed by the interior volume of a bank of several flat tubes, in which corrugated ribs coated with the conversion catalyst are placed.

3. A reactor as claimed in claim 2, wherein each of the flat tubes is made up of two tube half-shells, which are pre-fabricated to have a widening cross section towards the exterior and which are joined together in abutment around the periphery, being mechanically pre-stressed against an enclosed corrugated rib with a coating of the conversion catalyst at least on the rib flanks.

4. A reactor as claimed in claim 2 or 3, wherein the heat-exchanger structure is formed by a flat tube/rib block, in which several flat tubes forming a reaction chamber are arranged with heat-conductive corrugated ribs inserted in between on the outside of the flat tubes.

5. A reactor as claimed in claim 4, wherein the flat tubes are wider at the ends, the ends of adjacent flat tubes abutting against one another and being permanently joined to one another.

6. A reactor as claimed in claim 4 or 5, wherein the flat tube/tube block is inserted in a housing and the heat exchanger chamber on the outside of the flat tubes contains flow passages running in the longitudinal direction of its associated corrugated ribs, the housing having respective connecting structures for delivering and discharging heat exchange medium passing through the heat exchanger chamber on the outside of the flat tubes and the flow of operating substance directed through the reaction chamber.

7. A reactor as claimed in claim 6, wherein the heat exchanger chamber on the outside of the flat tubes is divided by an appropriately designed heat exchange medium-connecting structure into several part-compartments through which the heat exchange medium is passed in parallel or in series.

8. A reactor as claimed in claim 1, wherein the reaction chamber comprises a stack of corrugated ribs coated with the conversion catalyst and disposed crosswise one above the other and the stack of corrugated ribs has a bank of parallel heat exchange tubes, spaced at a distance apart, passing through it, the tube interior of which forms the second heat exchanger chamber for passing a heat exchange medium therethrough.

9. A reactor for catalytically converting a flow of operating substance, substantially as described herein with reference to the accompanying drawings.

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Claims searched: 1-9
Examiner: Chris Archer
Date of search: 18 January 2001

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Search Report under Section 17

Databases searched:

UK Patent Office collections, including GB, EP, WO & US patent specifications, in:

UK Cl (Ed.S): B1F (FD3, FD1B, FD1E, FD1X1)

Int Cl (Ed.7): B01J; F01N (3/28)

Other: ONLINE: WPI, EPODOC, JAPIO

Documents considered to be relevant:

Category	Identity of document and relevant passage		Relevant to claims
X	GB 1604980	(3M) see in particular Fig. 1	1
X	US 3910042	(YUGE ET AL) see in particular Fig. 2 and column 3 line 53 to column 4 line 24	1,2,4,6-8

X	Document indicating lack of novelty or inventive step	A	Document indicating technological background and/or state of the art.
Y	Document indicating lack of inventive step if combined with one or more other documents of same category.	P	Document published on or after the declared priority date but before the filing date of this invention.
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